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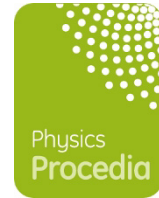
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Magnetic phases in $\text{Sr}_{1-x}\text{Ca}_x\text{Co}_2\text{P}_2$ studied by $\mu^+\text{SR}$

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Abstract

In order to elucidate the dependence of the magnetic ground state on the Ca content (x) in $\text{Sr}_{1-x}\text{Ca}_x\text{Co}_2\text{P}_2$ ($0 \leq x \leq 1$, ThCr_2Si_2 -type structure), we have performed muon spin rotation and relaxation ($\mu^+\text{SR}$) experiments on $\text{Sr}_{1-x}\text{Ca}_x\text{Co}_2\text{P}_2$ powder samples mainly in a zero applied field. The end member compound, SrCo_2P_2 , is found to be paramagnetic down to 19 mK. As x increases, such a paramagnetic ground state is observed down to 1.8 K until $x = 0.45$. Then, as x increases further, a short-range antiferromagnetic (AF) ordered phase appears at low temperatures for $0.48 \leq x \leq 0.75$, and finally, a long-range AF ordered phase is stabilized for $x > 0.75$. The internal magnetic field of the other end member compound, CaCo_2P_2 , is well consistent with that of the A -type AF order state, which was proposed from neutron scattering experiments. The phase diagram determined with $\mu^+\text{SR}$ is different from that proposed by macroscopic measurements. For an isostructural compound, LaCo_2P_2 , static magnetic order is found to be formed below ~ 130 K.

Keywords: muon spin rotation and relaxation, cobalt phosphide, antiferromagnet, phase diagram

1 Introduction

Although iron pnictides with the ThCr_2Si_2 -type (122) structure, e.g. CaFe_2As_2 and BaFe_2As_2 , show unconventional superconductivity under pressure and/or with substitution for Ba by K

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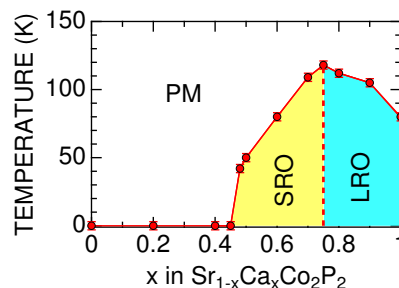


Figure 1: The variation of magnetic phases with x for $\text{Sr}_{1-x}\text{Ca}_x\text{Co}_2\text{P}_2$ determined with $\mu^+\text{SR}$ [8]. PM is a paramagnetic phase, SRO is a phase with wide field distribution probably due to short-range AF order, LRO is a long-range A -type AF ordered phase. Copyright 2015 American Physical Society.

[1, 2, 3], the related compounds AM_2P_2 with $A = \text{Ca, Sr, or Ba}$, and $M = \text{Fe, Co, or Ni}$ exhibit, instead, interesting properties with associated structural changes. For the present target system, $\text{Sr}_{1-x}\text{Ca}_x\text{Co}_2\text{P}_2$, the crystal structure changes from an uncollapsed tetragonal (ucT) phase for $x = 0$ to a collapsed tetragonal (cT) phase for $x = 1$ [4]. Based on macroscopic measurements [4], the system evolves from a nonmagnetic metallic ground state to an AF metallic ground state through a crossover composition regime at $x \sim 0.5$. Then, in the x range between 0.8 and 0.9, the system manifests a ferromagnetic (FM)-like ground state within which the magnetic ordering temperature is highest for $x \sim 0.9$. For the sample with $x \geq 0.9$, an AF ground state reappears [5]. Similar behavior was also reported for $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{P}_2$ [6], $\text{Ca}(\text{Ni}_{1-x}\text{Co}_x)_2\text{P}_2$ [6], and $\text{SrCo}_2(\text{Ge}_{1-x}\text{P}_x)_2$ [7].

However, according to our muon spin rotation and relaxation ($\mu^+\text{SR}$) experiment on $\text{Sr}_{1-x}\text{Ca}_x\text{Co}_2\text{P}_2$ [8] (Fig. 1), a paramagnetic ground state is stable in the x range between 0 and 0.45 down to the lowest temperature measured. Then, as x increases from 0.45, a short-range antiferromagnetic (AF) ordered phase appears at low temperatures for $0.48 \leq x \leq 0.75$, and finally, a long-range AF ordered phase is stabilized for $x > 0.75$. The discrepancy between the phase diagram obtained by macroscopic measurements and that by $\mu^+\text{SR}$ is due to the unique spatial and time resolution of $\mu^+\text{SR}$ [9, 10, 11, 12, 13].

Since the phase diagram of $\text{Sr}_{1-x}\text{Ca}_x\text{Co}_2\text{P}_2$ was already described in detail [8], here, we report the detailed $\mu^+\text{SR}$ result on SrCo_2P_2 , CaCo_2P_2 and an isostructural compound LaCo_2P_2 .

2 Experimental

Polycrystalline samples of SrCo_2P_2 , CaCo_2P_2 , and LaCo_2P_2 were prepared from elemental P, Sr, Ca, La, and Co using a two step reaction. For the first step, SrP, CaP, LaP, and Co_2P were synthesized by a solid state reaction between Sr (Ca, La, Co) and P in an evacuated quartz tube at 800°C (700°C for Co_2P). In the second step, SrCo_2P_2 , CaCo_2P_2 , and LaCo_2P_2 were synthesized by a solid state reaction between SrP, CaP, LaP, and Co_2P at 1000°C for 20 hours in an Ar atmosphere. After grinding, the obtained powder was annealed two times at 1000°C for 40 hours in an Ar atmosphere [14].

High quality single-crystal platelets of SrCo_2P_2 were grown by a flux technique using ele-

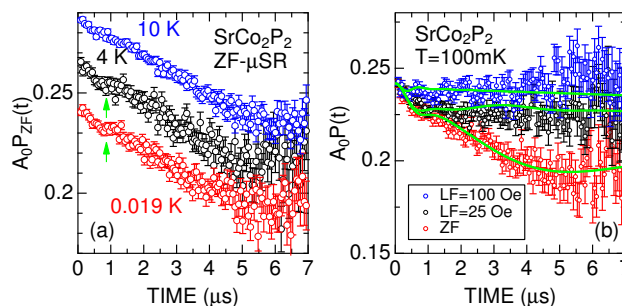


Figure 2: (a) The ZF- μ^+ SR time spectrum for $SrCo_2P_2$ measured at 19 mK, 4 K, and 10 K. Green arrows represent the small minimum appeared at $T \leq 4$ K. Each spectrum is shifted upward by 0.02 for clarity of display. (b) The ZF- and two LF- μ^+ SR spectra for $SrCo_2P_2$ measured at 100 mK. Solid lines in (b) represent the best fit with Eq. (1).

mental Sr, Co, and P as starting materials. Sn was used as the flux. The mixture of Sr, Co, P, and Sn were sealed in a quartz tube in an Ar atmosphere with 0.3 atm, and heated at 900°C for 72 hours, and then cooled down to 600°C with a rate of 3°C/h. The typical dimension of the crystal is $3 \times 3 \times 0.5$ mm³.

According to powder x-ray diffraction (XRD) analyses, all the samples were almost single phase of tetragonal symmetry with space group $I4/mmm$. The μ^+ SR spectra were measured at surface muon beam lines using the **LAMPF** spectrometer on M15 and M20 of TRIUMF in Canada and **Dolly** and **LTF** spectrometers of PSI in Switzerland. On **LAMPF** and **Dolly**, approximately 500 mg of powder sample was placed in an envelope with 1×1 cm² area, made of 0.05 mm thick Al-coated Mylar tape in order to minimize the background signal from the envelope. The envelope was attached to a low-background sample holder in a liquid-He flow-type cryostat for measurements in the T range between 1.8 and 150 K. At **LTF**, about 100 mg of platelets was attached onto a silver plate with an Apiezon-N grease, and the silver cell was set into a dilution refrigerator (DR) down to $T = 19$ mK. The experimental techniques are described in more detail elsewhere [9, 10].

3 Results and Discussion

3.1 $SrCo_2P_2$

Figure 2(a) shows the zero field (ZF-) μ^+ SR spectra for the $SrCo_2P_2$ crystals recorded at $T = 19$ mK, 4 K and 10 K. The ZF-spectrum exhibits a clear Kubo-Toyabe type relaxation at 10 K, indicating a paramagnetic nature of $SrCo_2P_2$. However, a small minimum appears around $t = 1 \mu s$ at 4 K and 19 mK, while the shape and position of the minimum does not depend on temperature at $T \leq 4$ K. Such behavior was also observed for a powder sample. In order to understand the nature of such minima, μ^+ SR spectra were also recorded under ZF and in two longitudinal fields (LF = 25 and 100 Oe). (Here, a longitudinal field means the field parallel to the initial muon spin polarization.) Figure 2(b) shows the ZF- and two LF- μ^+ SR spectra for the $SrCo_2P_2$ crystals obtained at 100 mK. The relaxation in the ZF-spectrum is clearly suppressed by LF, i.e. a decoupling behavior due to LF. The ZF- and LF-spectra

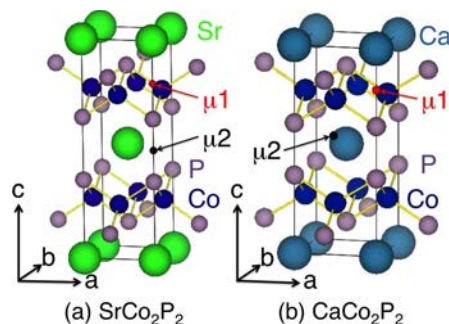


Figure 3: Muon sites in (a) SrCo_2P_2 and (b) CaCo_2P_2 deduced by DFT calculations [8]. The implanted muons are located at the $4e$ sites (μ_1 site) and $2b$ sites (μ_2 site) for SrCo_2P_2 and the $4e$ sites (μ_1 site) and $32o$ sites (μ_2 site) for CaCo_2P_2 . The atomic position of each site is given in Table 1. Copyright 2015 American Physical Society.

were well fitted by a combination of two static Kubo-Toyabe functions $[G(\Delta, t, H_{\text{LF}})]$ and a time-independent background signal from muons stopping in the silver plate.

$$A_0 P(t) = A_{\text{KT1}} G(\Delta_1, t, H_{\text{LF}}) + A_{\text{KT2}} G(\Delta_2, t, H_{\text{LF}}) + A_{\text{BG}}. \quad (1)$$

Here A_i are the asymmetries and Δ_i are the field distribution width at the muon sites. The fit for the ZF- and two LF-spectra using common A_{KT1} , A_{KT2} , A_{BG} , Δ_1 , and Δ_2 provided that $A_{\text{KT1}} = 0.0144 \pm 0.0011$, $A_{\text{KT2}} = 0.0364 \pm 0.0017$, $A_{\text{BG}} = 0.1910 \pm 0.0018$, $\Delta_1 = (2.33 \pm 0.09) \times 10^6 \text{ sec}^{-1}$, and $\Delta_2 = (0.337 \pm 0.014) \times 10^6 \text{ sec}^{-1}$. This means that there are two different muon sites in the SrCo_2P_2 lattice, and the muons at the two sites sense a small random internal magnetic field. Therefore, SrCo_2P_2 is found to be a paramagnet even at 19 mK, being consistent with the recent study on a single crystal down to 1.5 K [15]. In addition, a relatively large A_{BG} compared with A_{KT1} and A_{KT2} is due to a small sample size.

DFT calculations with generalized gradient approximation (GGA) [16] predicted the presence of two muon sites in the lattice (Fig. 3 and Table 1). The ratio between Δ s for the two sites is $6.9 (= 2.33/0.377)$ from the experiment, vs. $2.0 (= 0.4860/0.2425)$ from the calculations. Note that only nuclear magnetic moments were taken into account for the calculations. Since Δ_2 ranges between the two predicted values, the muons at one site, probably at the μ_2 site, clearly see a nuclear magnetic field. However, $\Delta_1 (= 2.33 \times 10^6 \text{ sec}^{-1} \sim 27 \text{ Oe})$ is rather large compared with the predicted values. This implies that the muons at the μ_1 site sense not only a nuclear magnetic field but also randomly distributed localized magnetic moments of Co, which appear below 10 K. The magnitude of such localized moment is estimated as $0.014 \mu_{\text{B}}$. Note that $A_{\text{KT1}}/(A_{\text{KT1}} + A_{\text{KT2}}) = 0.28$, corresponding to that 28 % muons in the SrCo_2P_2 sample see the internal magnetic field due to the Co moments. Therefore, such behavior is not induced by magnetic impurities but an intrinsic nature of SrCo_2P_2 and is likely to suggest the appearance of short-range correlation below 10 K. This is because ZF- and LF-spectra for the ordered phase is not explained by a static Kubo-Toyabe function [8].

Table 1: Possible muon sites (μ_n) and the field distribution width in SrCo_2P_2 and CaCo_2P_2 predicted by DFT calculations with GGA and dipole field calculations. The optimized structural parameters (and experimental values [5, 17]) are $a = 3.792$ (3.760) Å, $c = 11.793$ (11.602) Å, and $z = 0.3511$ (0.3525) for SrCo_2P_2 , and $a = 3.843$ (3.85) Å, $c = 9.603$ (9.55) Å, and $z = 0.3714$ (0.3722) for CaCo_2P_2 . Here, the atomic positions of Sr/Ca, Co, and P are (0,0,0), (0,1/2,1/4), and (0,0,z), respectively. Δ is calculated based only on nuclear magnetic moments and 1 Oe corresponds to $0.08516 \times 10^6 \text{ s}^{-1}$. E represents the potential energy. There are 4 equivalent positions in the unit cell for the 4e sites, 2 equivalent positions for the 2b sites, and 32 equivalent positions for the 32o sites. One of them for each site is shown in Fig. 3.

compound	site	(x, y, z)	E (eV)	Δ (Oe)	Δ (10^6 s^{-1})
SrCo_2P_2	4e (μ_1)	(0.0000,0.0000,0.1954)	-13.93	5.707	0.4860
	2b (μ_2)	(0.0000,0.0000,0.5000)	-14.10	2.848	0.2425
CaCo_2P_2	4e (μ_1)	(0.0000,0.0000,0.1991)	-14.10	5.912	0.5035
	32o (μ_2)	(0.5099,0.0253,0.0717)	-14.23	4.944	0.4210

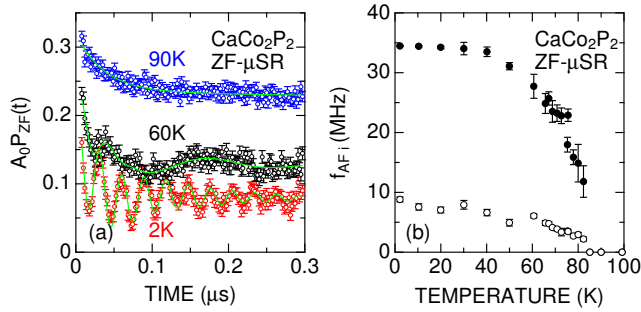


Figure 4: (a) the ZF- μ^+ SR spectra for CaCo_2P_2 obtained at 2, 60, and 90 K. Each spectrum is shifted upward by 0.05 for clarity of display. (b) the temperature dependence of the muon spin precession frequencies (f_{AFi}) in CaCo_2P_2 . Solid lines in (a) represent the best fit using Eq. (2).

3.2 CaCo_2P_2

Neutron diffraction studies [5] revealed that CaCo_2P_2 is an antiferromagnetic metal with $T_N \sim 85$ K, below which the Co moments (μ_{Co}) are aligned ferromagnetically in the c -plane, but antiferromagnetically along the c -axis below T_N , i.e. the A-type AF order with $\mathbf{q}=(0,0,1)$. The ZF- μ^+ SR spectrum for CaCo_2P_2 exhibits a clear oscillation due to the formation of static AF order below T_N . Since there are two oscillatory signals with different muon spin precession frequencies, the ZF spectrum was fitted by

$$A_0 P_{\text{ZF}}(t) = \sum_{i=1}^2 A_{\text{AF}i} \cos(2\pi f_{\text{AF}i} t + \phi_i) \exp(-\lambda_{\text{AF}i} t) + A_{\text{tail}} \exp(-(\lambda_{\text{tail}} t)^\gamma) + A_{\text{BG}} G(\Delta_{\text{BG}}, t), \quad (2)$$

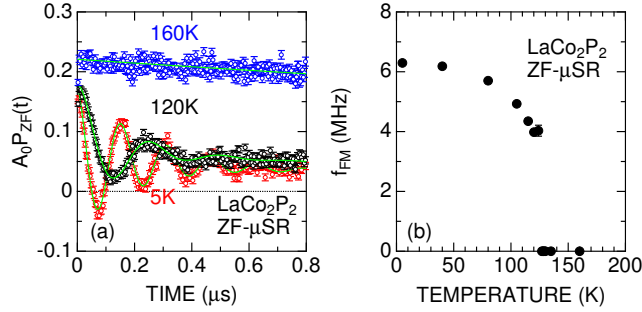


Figure 5: (a) the ZF-μ⁺SR spectra for LaCo₂P₂ obtained at 5, 120, and 160 K. (b) the temperature dependence of the muon spin precession frequency (f_{FM}) in LaCo₂P₂. Solid lines in (a) represent the best fit using Eq. (3).

where A_i are the asymmetries and $A_{tail} = \frac{1}{2} \sum A_{AFi}$, λ_i are exponential relaxation rates, and $2\pi f_i (\equiv \omega_i)$ are the muon Larmor frequencies of the three signals. G is a static Kubo-Toyabe function [18], $G = \frac{1}{3} + \frac{2}{3}(1 - \Delta^2 t^2) \exp(-\frac{1}{2} \Delta^2 t^2)$, and Δ (in this case Δ_{BG}) is proportional to the field distribution width at the muon site. The A_{tail} signal corresponds to the ZF “1/3 tail” due to the field component parallel to the initial muon spin polarization. Finally, the A_{BG} signal corresponds to a static nuclear field component due to a nonmagnetic impurity phase with about 13 vol%, most likely CaP. In fact, $\Delta_{BG} = 0.358(8) \times 10^6 \text{ s}^{-1}$ (implying a distribution of local fields with a width of $\sim 4.2 \text{ Oe}$) at 2 K and was almost T independent until T_N .

Both $f_{AF1}(T)$ and $f_{AF2}(T)$ curves show an order parameter like T dependence and drop to zero at T_N [see Fig. 4(b)], indicating the presence of two magnetically different muon sites in the lattice, as predicted by DFT calculations [Fig. 3(b) and Table 1]. If we compare the internal magnetic field estimated by dipole field calculations at the two muon sites with the experimental results, we obtain that $\mu_{Co} = 0.41(1) \mu_B$ at 2 K, which is roughly comparable to the past neutron work ($0.32 \mu_B$) [5]. Such small μ_{Co} compared with the effective magnetic moment of Co ($\mu_{eff} \sim 1.7 \mu_B$ [4]) supports an itinerant electron nature of CaCo₂P₂.

3.3 LaCo₂P₂

LaCo₂P₂ is known as a ferromagnetic metal with $T_C = 132 \text{ K}$ [19]. Figure 5(a) shows the variation of the ZF-μ⁺SR spectrum with temperature. Since the spectrum below T_C exhibits a clear oscillation due to the formation of a static magnetic field, the spectrum was fitted by a combination of an exponentially damped cosine oscillation for the static internal magnetic field and an exponentially damped non-oscillatory signal for a “1/3” tail for a powder sample;

$$A_0 P_{ZF}(t) = A_{FM} \exp(-\lambda_{FM} t) \cos(2\pi f_{FM} t + \phi_{FM}) + A_{tail} \exp(-\lambda_{tail} t), \quad (3)$$

where A_i are the asymmetries, λ_i are the exponential relaxation rates, f_{FM} is the muon spin precession frequency, and ϕ_{FM} is the initial phase.

Figure 5(b) shows the temperature dependence of f_{FM} for LaCo₂P₂. The $f_{FM}(T)$ curve exhibits an order parameter-like temperature dependence, as expected. However, for unmagnetized ferromagnetic materials in zero applied field, the internal magnetic field at a muon site

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(\mathbf{H}_μ) is represented by [20, 21, 22];

$$\mathbf{H}_\mu = \mathbf{H}_{\text{dip}} + \mathbf{H}_\mathbf{L} + \mathbf{H}_{\text{hf}}, \quad (4)$$

where \mathbf{H}_{dip} is the dipolar field, $\mathbf{H}_\mathbf{L}$ is the Lorentz field, and \mathbf{H}_{hf} is the hyperfine field. Furthermore, $\mathbf{H}_\mathbf{L}$ and \mathbf{H}_{hf} are connected to the saturated magnetization ($\mathbf{M}_\mathbf{s}$) and the local spin density at the muon sites (ρ_{spin}), as follows;

$$\begin{aligned} \mathbf{H}_\mathbf{L} &= 4\pi/3 \times \mathbf{M}_\mathbf{s}, \\ \mathbf{H}_{\text{hf}} &= 8\pi/3 \times \rho_{\text{spin}}(\mathbf{r}_\mu). \end{aligned} \quad (5)$$

Since $M_\mathbf{s} = 0.391 \mu_\text{B}/\text{Co}$ along the a -axis at 5 K [19], $\mathbf{H}_\mathbf{L}$ is calculated as (190, 0, 0) Oe [22].

If we assume that the implanted muons locate at the $\mu 1$ site, as in the case for SrCo_2P_2 , and that the Co moments align along the a -axis, as proposed by neutron diffraction measurements, then $H_{\text{dip}} = 1884 \text{ Oe}/\mu_\text{B}$ along the a -axis. Since the ordered moment was reported as $\mu_{\text{ord}} = 0.47(5) \mu_\text{B}$, $\mathbf{H}_{\text{dip}} = (885, 0, 0) \text{ Oe}$ and $\mathbf{H}_{\text{hf}} = (-610, 0, 0) \text{ Oe}$. Thus, \mathbf{H}_{hf} is found to be almost comparable to \mathbf{H}_{dip} , as in the case of other ferromagnetic materials [22, 23, 24].

4 Summary

The magnetic phase diagram of the solid solution system between SrCo_2P_2 and CaCo_2P_2 was studied with $\mu^+\text{SR}$ using mainly powder samples. The end compound, SrCo_2P_2 , was found to be an enhanced paramagnetic metal [25] down to 19 mK, although small random localized magnetic moments, which probably suggests the presence of short-range correlation, appear below 10 K. The other end compound, CaCo_2P_2 , enters into an A -type antiferromagnetic ordered phase below ~ 85 K. The isostructural compound, LaCo_2P_2 , was found to undergo a magnetic transition below ~ 130 K. Using the past neutron data, the hyperfine field was also estimated.

5 Acknowledgments

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